Fundamental Physics with Two Radioactive Molecules

Timo Fleig

LCPQ, FeRMI
Université Paul Sabatier Toulouse III
France

26 March 2024

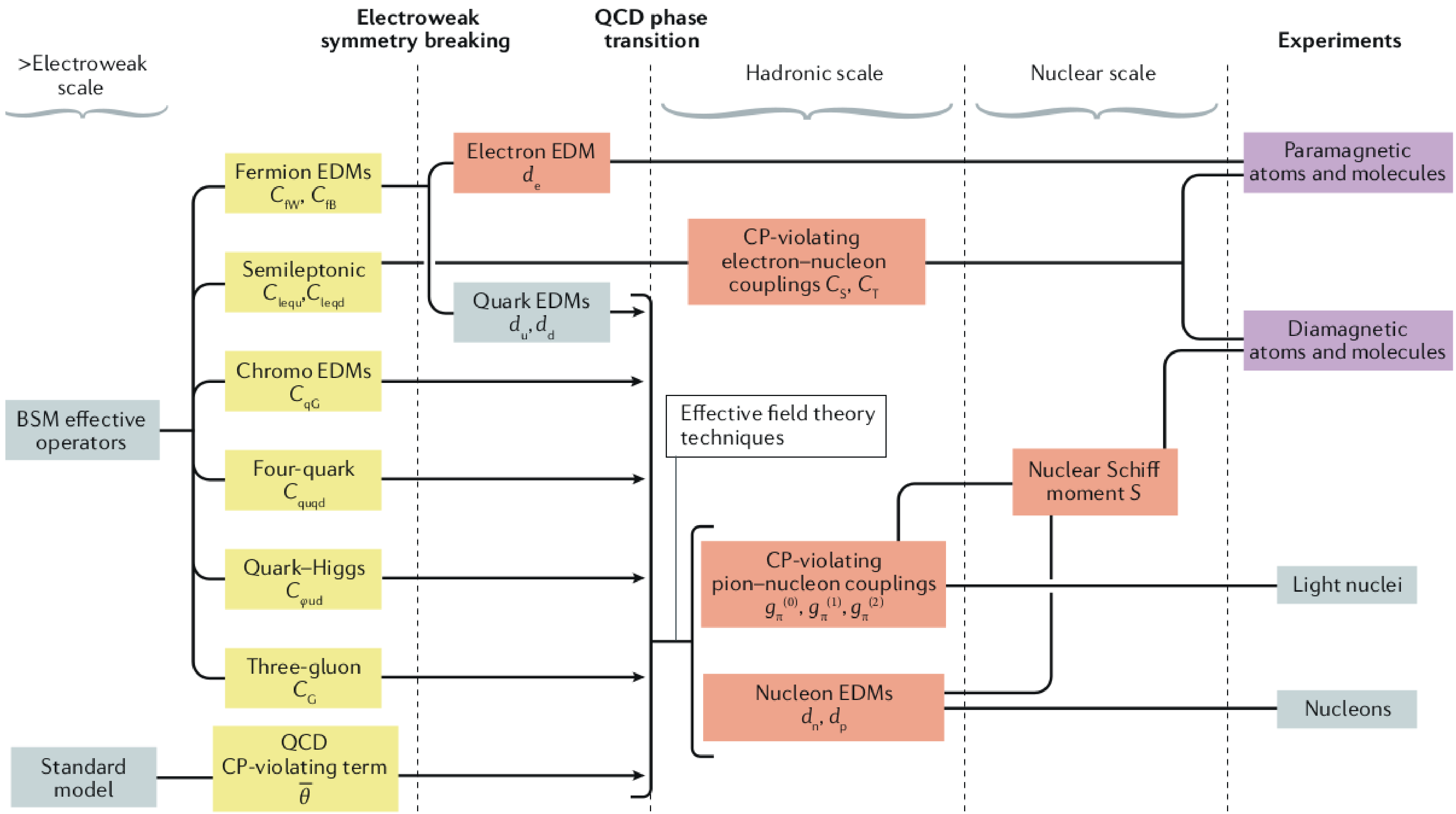
Outline

Hadron-sector searches: **Schiff-moment interaction**

Search for a lepton EDM: **Electron EDM interaction**

Hadron-sector searches: **Tensor-pseudotensor interaction**

EDMs and their possible sources: An overview



W. Cairncross, J. Ye, *Nat. Rev. Phys.* **1** (2019) *510*

EDM Science

- Electron EDM interactions (HfF^+, ThO, Hg, Tl, TaO^+, RaAg et al.)

  T. F., D. DeMille, *New J. Phys.* **23** (2021) *113039*

  T. F., L. V. Skripnikov, *Symmetry* **12** (2020) *498*

  T. F., M. Jung, *J High Energy Phys. (JHEP)* **07** (2018) *012*

  T. F., *Phys. Rev. A* **96** (2017) *040502(R)*

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  M. Denis, T. F., *J. Chem. Phys.* **145** (2016) *214307*

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Atomic and Molecular Correlated Wavefunctions[1]

Hamiltonians

- Dirac-Coulomb Hamiltonian + external electric field (atoms)

$$\hat{H}^{\text{Dirac-Coulomb}} + \hat{H}^{\text{Int-Dipole}} = \sum_i^n \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 - \frac{Z}{r_i} 1\!\!1_4 \right] + \sum_{i,j>j}^n \frac{1}{r_{ij}} 1\!\!1_4 + \sum_i^n \mathbf{r}_i \cdot \mathbf{E}_{\text{ext}}\, 1\!\!1_4$$

- Dirac-Coulomb Hamiltonian operator (molecules)

$$\hat{H}^{DC} = \sum_i^n \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 - \sum_A^N \frac{Z}{r_{iA}} 1\!\!1_4 \right] + \sum_{i,j>i}^n \frac{1}{r_{ij}} 1\!\!1_4 + \sum_{A,B>A}^N V_{AB}$$

- Dirac-Coulomb-Gaunt[2] Hamiltonian operator (molecules)

$$\hat{H}^{DCG} = \sum_i^n \left[c\,\boldsymbol{\alpha}_i \cdot \mathbf{p}_i + \beta_i c^2 - \sum_A^N \frac{Z}{r_{iA}} 1\!\!1_4 \right] + \sum_{i,j>i}^n \left(\frac{1}{r_{ij}} 1\!\!1_4 - \frac{1}{2} \frac{\vec{\alpha}_i \vec{\alpha}_j}{r_{ij}} \right) + \sum_{A,B>A}^N V_{AB}$$

---

[1] T. F., H.J.Å. Jensen, J. Olsen, L. Visscher, *J Chem Phys* **124** (2006) *104106*
S. Knecht, H.J.Å. Jensen, T. F., *J Chem Phys* **132** (2010) *014108*

[2] A. Marc, T.F., in preparation

Calculation of Properties Including $\mathcal{P}$,$\mathcal{T}$-Violating Effects[3]

Using String-Based CI Techniques

Solve CI problem $\Rightarrow \psi_k^{(0)}$; expectation value over relativistic Configuration Interaction wavefunction

$$\left\langle \hat{O} \right\rangle_{\psi_k^{(0)}} = \sum_{I,J=1}^{\mathrm{dim}\mathcal{F}^{\mathrm{t}}(\mathrm{M,n})} c_{kI}^* c_{kJ} \; \langle \mid (\mathcal{S}\overline{\mathcal{T}})_I^\dagger \left| \hat{O} \right| (\mathcal{S}\overline{\mathcal{T}})_J \mid \rangle$$

Property operator $\hat{O}$ in basis of Kramers-paired atomic/molecular spinors

$$\hat{O} = \sum_{i,j=1}^{P_u} o_{ij}\, a_i^\dagger a_j + \sum_{i=1}^{P_u} \sum_{j=P_u+1}^{P} o_{i\overline{j}}\, a_i^\dagger a_{\overline{j}} + \sum_{i=P_u+1}^{P} \sum_{j=1}^{P_u} o_{\overline{i}j}\, a_{\overline{i}}^\dagger a_j + \sum_{i,j=P_u+1}^{P} o_{\overline{ij}}\, a_{\overline{i}}^\dagger a_{\overline{j}}$$

First-term contribution to expectation value

$$O(\Psi_k)_1 = \sum_{I,J=1}^{\mathrm{dim}\mathcal{F}^{\mathrm{t}}(\mathrm{M,n})} c_{kI}^* c_{kJ} \sum_{i,j=1}^{P_u} o_{ij}$$
$$\langle \mid \prod_{p=1}^{N_p} \prod_{\overline{p}=N_p+1}^{N_p+N_{\overline{p}}} a_{\overline{p}} a_p \, a_i^\dagger a_j \prod_{q=1}^{N_p} \prod_{\overline{q}=N_p+1}^{N_p+N_{\overline{p}}} a_q^\dagger a_{\overline{q}}^\dagger \mid \rangle$$

[3] S. Knecht, Dissertation, HHU Düsseldorf (2009)
T. F., M.K. Nayak, *Phys Rev A* **88** (2013) *032514*

Atomic EDM

in terms of underlying symmetry breaking

Electric dipole moment of an atom:[4]

$$d_a := -\lim_{E_{\text{ext}}\to 0}\left[\frac{\partial(\Delta\varepsilon_{\not{P}\not{T}})}{\partial E_{\text{ext}}}\right]$$

$\Delta\varepsilon_{\not{P}\not{T}}$ is **some** P,T-odd energy shift.

Sources are particle EDMs, nuclear MQM, nuclear Schiff moment, $\mathcal{T}$-odd contribution to weak interaction etc.

For some Hamiltonian $\hat{H}_{\not{P}\not{T}} = \alpha\hat{O}_{\not{P}\not{T}}$, we then have

$$d_a = -\lim_{E_{\text{ext}}\to 0}\frac{\partial}{\partial E_{\text{ext}}}\alpha\left\langle\hat{O}_{\not{P}\not{T}}\right\rangle_{\psi(E_{\text{ext}})}$$

Defining a general interaction constant as $R := \frac{d_a}{\alpha}$ the linear-regime atomic interaction constant is then:

$$R = -\lim_{E_{\text{ext}}\to 0}\frac{\left\langle\hat{O}_{\not{P}\not{T}}\right\rangle_{\psi(E_{\text{ext}})}}{E_{\text{ext}}} \approx R_{\text{lin}} = -\frac{\left\langle\hat{O}_{\not{P}\not{T}}\right\rangle_{\psi(E_{\text{ext}})}}{E_{\text{ext}}}$$

with finite but very small E_{ext}.

[4] E.D. Commins, *Adv. Mol. Opt. Phys.* **40** (1999) *1*

Search for Hadron-Source EDM: Schiff-Moment Interaction

Xe atom

Atomic interaction constant[5] as implemented into KRCI/DIRAC[6]

$$\alpha_{\mathrm{SM}} := \frac{\Delta\varepsilon_{\mathrm{SM}}}{S_z\, E_{\mathrm{ext}}} = \frac{-\frac{3}{B}\left\langle \sum_{j=1}^{n} \hat{z}_j\, \rho(\mathbf{r}_j) \right\rangle_{\psi(E_{\mathrm{ext}})}}{E_{\mathrm{ext}}}$$

DCHF	α_{SM} $\left[10^{-17}\frac{e\mathrm{cm}}{e\mathrm{fm}^3}\right]$	$\varepsilon_{\mathrm{DCHF}}$ [a.u.]
DZ-21s15p	-1.22	-7446.876435682
Dzuba *et al.* [5] (RPA, 2002)	0.38	-

[5] V. A. Dzuba, V. V. Flambaum, J. S. M. Ginges, and M. G. Kozlov, *Phys. Rev. A* **66** (2002) *021111*
M. Hubert, T.F., *Phys. Rev. A* **106** (2022) *022817*

[6] S. Knecht and H. J. Aa. Jensen, T.F., *J. Chem. Phys.* **132** (2010) *014108*
T. Saue *et al.*, *J. Chem. Phys.* **152** (2020) *204104*

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"Even-tempered" densification:

$$\frac{\zeta_{n-1}}{\zeta_n} := \frac{\zeta_n}{\zeta_{n+1}} \Leftrightarrow \zeta_n = \sqrt{\zeta_{n-1}\,\zeta_{n+1}}$$

[6] V. A. Dzuba, V. V. Flambaum, J. S. M. Ginges, and M. G. Kozlov, *Phys. Rev. A* **66** (2002) *021111*
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sp-densified+1sp QZ-69s57p	0.37	-7446.895401869
Dzuba *et al.* [6] (RPA, 2002)	0.38	-

[6] V. A. Dzuba, V. V. Flambaum, J. S. M. Ginges, and M. G. Kozlov, *Phys. Rev. A* **66** (2002) *021111*
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sp-densified+1sp QZ-69s57p	0.37	-7446.895401869
sp-densified+2sp QZ-71s59p	0.38	-7446.895401810
sp-densified+3sp QZ-73s61p	0.38	-7446.895401761
Dzuba *et al.* [6] (RPA, 2002)	0.38	-

[6] V. A. Dzuba, V. V. Flambaum, J. S. M. Ginges, and M. G. Kozlov, *Phys. Rev. A* **66** (2002) *021111*
M. Hubert, T.F., *Phys. Rev. A* **106** (2022) *022817*

Search for Hadron-Source EDM: Francium-Silver (FrAg)

Atomic Basis Sets / Hartree-Fock Theory

Molecular Schiff-moment interaction Hamiltonian[7]

$$W_{\mathrm{SM}}(A) := \frac{\Delta\varepsilon_{\mathrm{SM}}(A)}{S_z(A)} = -\frac{3}{B}\left\langle \sum_{j=1}^{n} \hat{z}_j\, \rho_A(\mathbf{r}_j) \right\rangle_{\psi}$$

as implemented into KRCI/DIRAC[8]

	CsLi ($R_e = 6.927$ a.u.)		FrAg ($R_e = 6.190$ a.u.)	
Basis	$\varepsilon_{\mathrm{DCHF}}$ [a.u.]	W_{SM} [a.u.]	$\varepsilon_{\mathrm{DCHF}}$ [a.u.]	W_{SM} [a.u.]
cvDZ	-7794.1925854	-10110	-29622.7980959	5946
cvTZ	-7794.2033064	-2849	-29622.8345496	28173
cvQZ	-7794.2038442	2098	-29622.8362766	29451
cvQZ+	-7794.2038394	2887	-29622.8354238	31350

[7] V. A. Dzuba, V. V. Flambaum, J. S. M. Ginges, and M. G. Kozlov, *Phys. Rev. A* **66** (2002) *021111*

[8] M. Hubert, T.F., *Phys. Rev. A* **106** (2022) *022817*

Search for Hadron-Source EDM: Francium-Silver (FrAg)

Electron Correlation Effects

Basis/cutoff	ε_{CI} [a.u.]	W_{SM} [a.u.]
cvQZ+/DCHF	-29622.8354238	31350
cvQZ+/SD2_2au	-29622.8604657	30359
cvQZ+/SD2_5au	-29622.8605445	30355
cvQZ+/SD2_8au	-29622.8605500	30360
cvQZ+/SD10_8au	-29623.0196812	29980
cvQZ+/SDT10_8au	-29623.0260848	29909
cvQZ+/SD12_8au	-29623.1920759	30711
cvQZ+/SD20_8au	-29623.3371101	30127
cvQZ+/SD36_5au	-29623.7102434	30333
cvQZ+/SD36_8au	-29623.8379481	30239

- SD36 model includes Fr$(7s, 6p, 6s, 5d)$ and Ag$(5s, 4d, 4p)$ shells
- Excitations out of Fr(s, p) shells diminish W_{SM}.
- Excitations out of other shells increase W_{SM}.

Search for Hadron-Source EDM: Francium-Silver (FrAg)

Comparison with other $^1\Sigma_0$ molecules

	Z (heavy)	EA (light) [eV]	W_{SM} [a.u.] (at respective R_e)
CsAg	55	1.304	3530[9]
FrLi	87	0.618	24414[9]
FrAg	87	1.304	30168 ± 2504[9]
TlF	81	3.401	39967 ± 3600[10]
			37192[11]
			40539[12]

- Cs $\longrightarrow$ Fr order of magnitude gain
- Li $\longrightarrow$ Ag substantial gain
- TlF benefits from huge EA(F)

[9] A. Marc, M. Hubert, T. F., *Phys. Rev. A* **108** (2023) *062815*

[10] M. Hubert, T.F., *Phys. Rev. A* **106** (2022) *022817*

[11] L. V. Skripnikov, N. S. Mosyagin, A. V. Titov, and V. V. Flambaum, *Phys. Chem. Chem. Phys.* **22** (2020) *18374*

[12] V. V. Flambaum, V. A. Dzuba, and H. B. Tran Tan, *Phys. Rev. A* **101** (2020) *042501*
A. N. Petrov, N. S. Mosyagin, T. A. Isaev, A. V. Titov, V. F. Ezhov, E. Eliav, and U. Kaldor, *Phys. Rev. Lett.* **88** (2002) *073001*

Search for Hadron-Source EDM: Francium-Silver (FrAg)

Schiff moment in terms of QCD $\overline{\theta}$ and π-meson–nucleon CP-violating interaction constants[13]

$$S\left({}^{223}\mathrm{Fr}\right) \approx \left(-4.16 g\bar{g}_0 + 20.64 g\bar{g}_1 - 11.04 g\bar{g}_2\right)\, e\mathrm{fm}^3$$
$$S\left({}^{205}\mathrm{Tl}\right) \approx \left(0.13\, g\bar{g}_0 - 0.004\, g\bar{g}_1 - 0.27\, g\bar{g}_2\right)\, e\mathrm{fm}^3$$
$$S\left({}^{225}\mathrm{Ra}\right) \approx \left(-2.6\, g\bar{g}_0 + 12.9\, g\bar{g}_1 - 6.9\, g\bar{g}_2\right)\, e\mathrm{fm}^3$$

$$S\left({}^{223}\mathrm{Fr}\right) \approx -1.6\,\overline{\theta} e\ \mathrm{fm}^3$$
$$S\left({}^{205}\mathrm{Tl}\right) \approx 0.02\,\overline{\theta} e\mathrm{fm}^3$$
$$S\left({}^{225}\mathrm{Ra}\right) \approx -\overline{\theta} e\mathrm{fm}^3$$

^{223}Fr orders of magnitude more sensitive than ^{205}Tl

[13] V. V. Flambaum, V. A. Dzuba, and H. B. Tran Tan, *Phys. Rev. A* **101** (2020) *042501*
V. A. Dzuba, V. V. Flambaum, *Phys. Rev. A* **101** (2020) *042504*

Current World Records

In the presence of a non-zero EDM d, the system's Hamiltonian is
$\hat{H} = -(\mu\mathbf{B} + d\mathbf{E}) \cdot \frac{\hat{\mathbf{J}}}{|J|}$

- **"Paramagnetic" systems**: Precession measurement on **HfF^+**
  JILA group; Ye, Cornell[14]
  measured $f = (-14.6 \pm 29.7)\ \mu$Hz $\Rightarrow |d_e| \leq 4.1 \times 10^{-30}\, e$ cm
- **"Diamagnetic" systems**: Precession measurement on **Hg**
  Seattle group; Heckel[15]
  measured $|d_{Hg}| \leq 7.4 \times 10^{-30}\, e$ cm
- **Neutron** (n) EDM experiment
  PSI, Switzerland[16]
  measured $|d_n| \leq 1.8 \times 10^{-26}\, e$ cm

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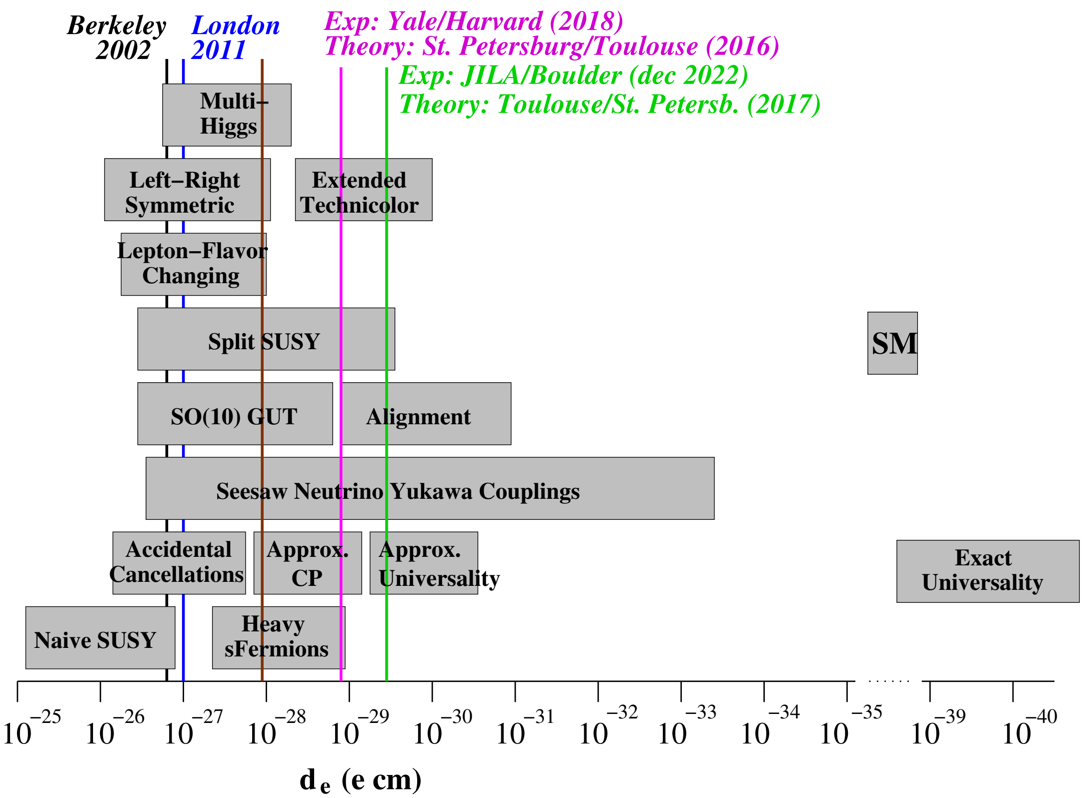
[14] T. S. Roussy, *et al.*, J. Ye, E. A. Cornell, Science **381** (2023) *46*

[15] B. Graner *et al.*, Phys Rev Lett **116** (2016) *161601*

[16] C. Abel *et al.*, Phys. Rev. Lett., **124** (2020) *081803*

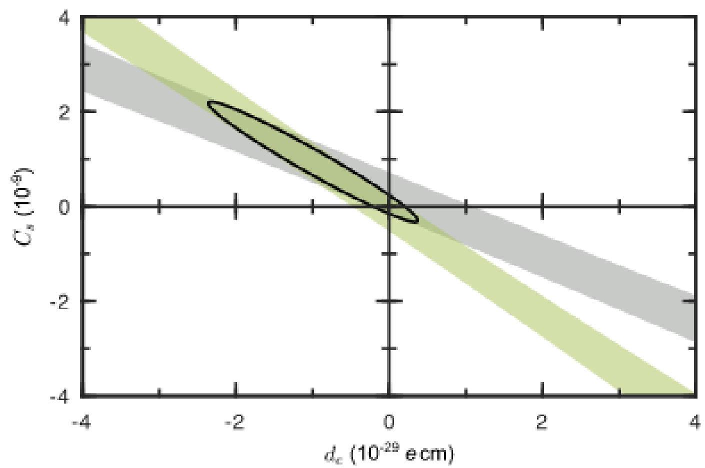
Updates: eEDM Constraint on BSM Theories (2023)



E_{eff} $\left[\frac{\text{GV}}{\text{cm}}\right]$

22.7[17] 22.5[18]

$|d_e| < 4.1 \times 10^{-30}\,e\text{cm}$ (90% C.L.)[19]



Combination with ThO measurement[20]:

$|d_e| < 2.1 \times 10^{-29}\,e\text{cm}$ (90% C.L.)[19]

$|C_S| < 1.9 \times 10^{-9}$ (90% C.L.)[19]

[17] T. F., *Phys. Rev. A* **96** (2017) *040502(R)*

[18] L. V. Skripnikov, *J. Chem. Phys.* **147** (2017) *021101*

[19] T. S. Roussy, L. Caldwell, T. Wright, W. B. Cairncross, Y. Shagam, K. B. Ng, N. Schlossberger, S. Y. Park, A. Wang, J. Ye, E. A. Cornell, *Science* **381** (2023) *46*

[20] ACME Collaboration, *Nature* **562** (2018) *355*

HfF^+ Electronic Structure and eEDM Interaction Constant[21]

GAS-CI definitions

Expectation value in many-body system in accord with stratagem II[21]

$$-\left\langle \sum_{j=1}^{n} \gamma_j^0\, \mathbf{\Sigma}_j \cdot \mathbf{E}_j \right\rangle_{\psi^{(0)}} \approx \frac{2\imath c}{e\hbar} \left\langle \sum_{j=1}^{n} \gamma_j^0 \gamma_j^5\, \mathbf{p}_j^{\;2} \right\rangle_{\psi^{(0)}} := E_{\mathrm{eff}}$$

- Basis: uncontracted vTZ
  Hf: $\{30s, 24p, 15d, 10f, 3g, 1h\}$
  F: $\{10s, 5p, 2d, 1f\}$
- Dirac-Coulomb Hamiltonian
- Full $(SS| **)$ integrals (EDM)

	# of Kramers pairs	accumulated # of electrons min.	accumulated # of electrons max.
Deleted	(164)		
Virtual	118	34	34
Hf: 6s, 5d	6	34–p	34
F: 2s, 2p	4	32–(m+n)	32
Hf: 5s, 5p F: 1s	5	24–m	24
Hf: 4f	7	14–q	14
Frozen core	(23)		

[21] T.F., *Phys. Rev. A* **96** (2017) *040502(R)*
[21] E. Lindroth, E. Lynn, P.G.H. Sandars, *J Phys B: At Mol Opt Phys* **22** (1989) *559*

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  M. Denis, T. F., *J. Chem. Phys.* **145** (2016) *214307*

- Nuclear Schiff-moment interactions (Xe, Hg, TlF, FrAg et al.)

  A. Marc, M. Hubert, T. F., *Phys. Rev. A* **108** (2023) *062815*

  M. Hubert, T. F., *Phys. Rev. A* **106** (2022) *022817*

- Weak neutral current interactions (Xe, Hg, Ra, TlF)

  T. F., *Phys. Rev. A* **109** (2024) *022807*

  T. F., *Phys. Rev. A* **99** (2019) *012515*

- Nuclear MQM interactions (TaN, TaO^+, HfF^+, RaAg)

  T. F., M. K. Nayak, M. G. Kozlov, *Phys. Rev. A* **93** (2016) *012505*

Search for the Electron EDM: Radium-Silver (RaAg)

in collaboration with

David DeMille

University of Chicago

Olivier Grasdijk

ARGONNE Labs / University of Chicago

Going Ultracold: From beams to traps

PHYSICAL REVIEW A, VOLUME 63, 023405

Loading and compressing Cs atoms in a very far-off-resonant light trap

D. J. Han, Marshall T. DePue, and David S. Weiss
Department of Physics, University of California at Berkeley, Berkeley, California 94720-7300
(Received 25 May 2000; published 12 January 2001)

We describe an experiment in which 3×10^7 Cs atoms are loaded into a 400 μm crossed beam far-off-resonant trap (FORT) that is only 2 μK deep. A high-density sample is prepared in a magneto-optic trap, cooled in a three-dimensional far-off-resonant lattice (FORL), optically pumped into the lowest-energy state, adiabatically released from the FORL, magnetically levitated, and transferred to the final trap with a phase-space density of 10^{-3}. Spontaneous emission in the FORT is negligible, and we have compressed the atoms in the FORT to a spatial density of 2×10^{13} atoms/cm^3. Evaporative cooling under these conditions proceeds rapidly.

- Estimated sensitivity of Cs EDM measurement in DLT[22] is $|d_e| \approx 10^{-29}\, e\text{cm}$

Cs atom: $\Delta E = R\, E_{\text{ext}}\, d_e$
$E_{\text{int}} \approx 20\ \left[\frac{\text{MV}}{\text{cm}}\right]$

Ultracold **XY** Molecule: $\Delta E = E_{\text{eff}}\, d_e$
$E_{\text{eff}} \approx 50\ \left[\frac{\text{GV}}{\text{cm}}\right]$

- A factor of $\approx \mathbf{2500}$ gain in sensitivity!

[22] DLT: Dipole light trap; D. Weiss (Penn State), 2014: "Measuring the eEDM using laser-cooled Cs atoms in optical lattices"
S. Chu, J.E. Bjorkholm, A. Ashkin, A. Cable, *Phys. Rev. Lett.* **57** (1986) *314*
C. Chin, V. Leiber, V. Vuletić, A.J. Kerman, S. Chu, *Phys. Rev. A* **63** (2001) *033401*

Towards Ultracold DLT EDM Measurement[23]

Picking the cherry

Target atom:

Z(Ra) $= 88$ α_D(Ra) $= 246 \pm 4$ a.u.[24]

Polarizing partner:

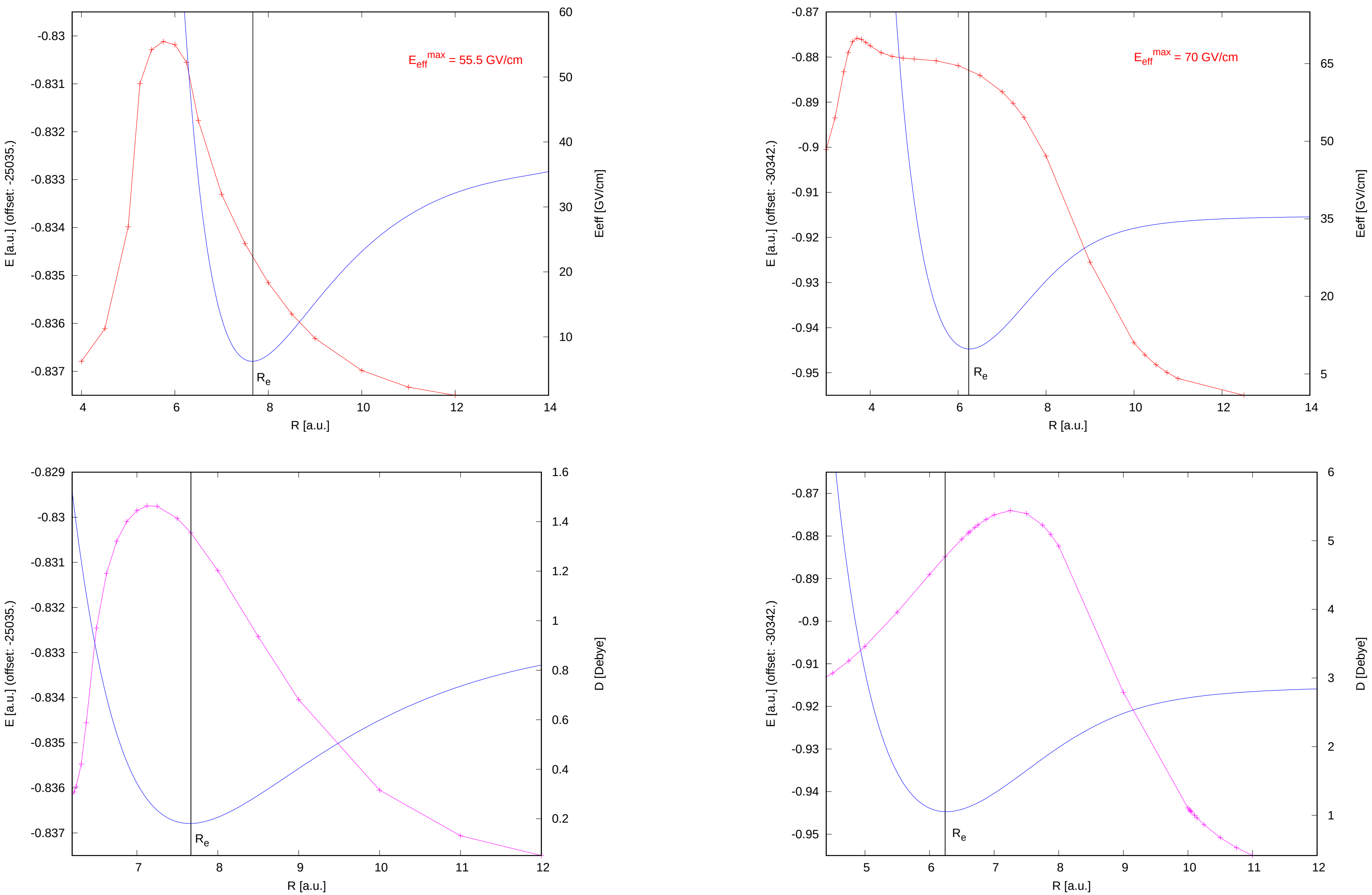
Alkali(-like) atoms: Li, Na, K, Rb, Cs, Fr; Cu, Ag, Au

	R_e [a.u.]	B_e [cm^{-1}]	D[Debye]	EA [eV]	E_{eff} $\left[\frac{\text{GV}}{\text{cm}}\right]$	W_S [kHz]	E_{pol} $\left[\frac{\text{kV}}{\text{cm}}\right]$
RaLi	7.668	0.151	1.36	0.618	22.2	-59.5	13.3
RaNa	8.703	0.038	0.51	0.548	12.0	-32.2	8.90
RaK	10.37	0.017	0.39	0.501	5.44	-14.6	5.18
RaRb	10.75	0.008	0.36	0.486	5.01	-13.6	2.75
RaCs	11.25	0.006	0.46	0.472	4.52	-12.6	1.48
RaFr	11.26	0.004	0.24	0.486	3.44	-12.4	2.06
RaCu	6.050	0.033	**4.30**	**1.236**	67.0	-180.6	0.92
RaAg	6.241	0.021	**4.76**	**1.304**	**63.9**	**-175.1**	0.53
RaAu	5.836	0.017	**5.71**	**2.309**	50.4	-166.4	0.36

[23] T. F., D. DeMille, *New J. Phys.* **23** (2021) *113039*

[24] P. Schwerdtfeger, J. K. Nagle, *Mol. Phys.* **117** (2019) *1200*

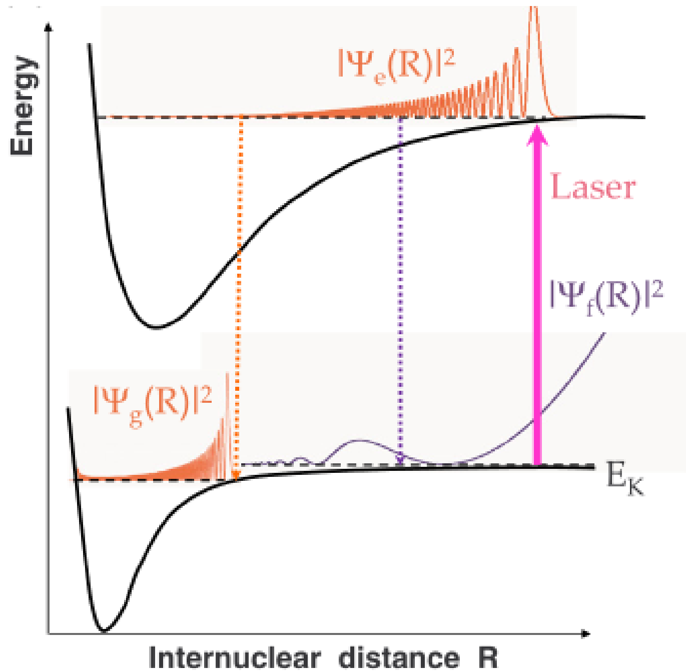
RaLi vs. RaAg[25]



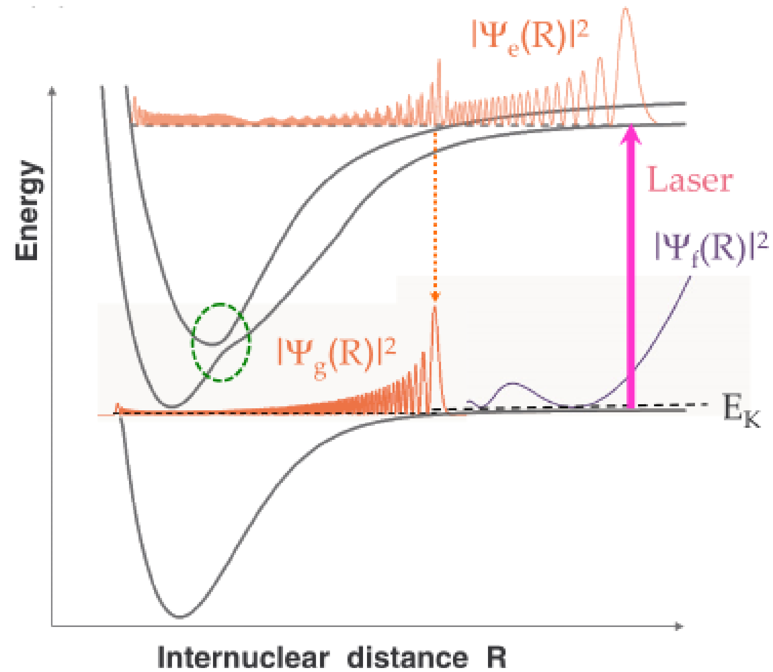
[25] T. F., D. DeMille, *New J. Phys.* **23** (2021) *113039*

"Building" RaAg in a DLT EDM Experiment

- Photoassociating ultracold atoms into ultracold molecules[26]



1) Direct approach

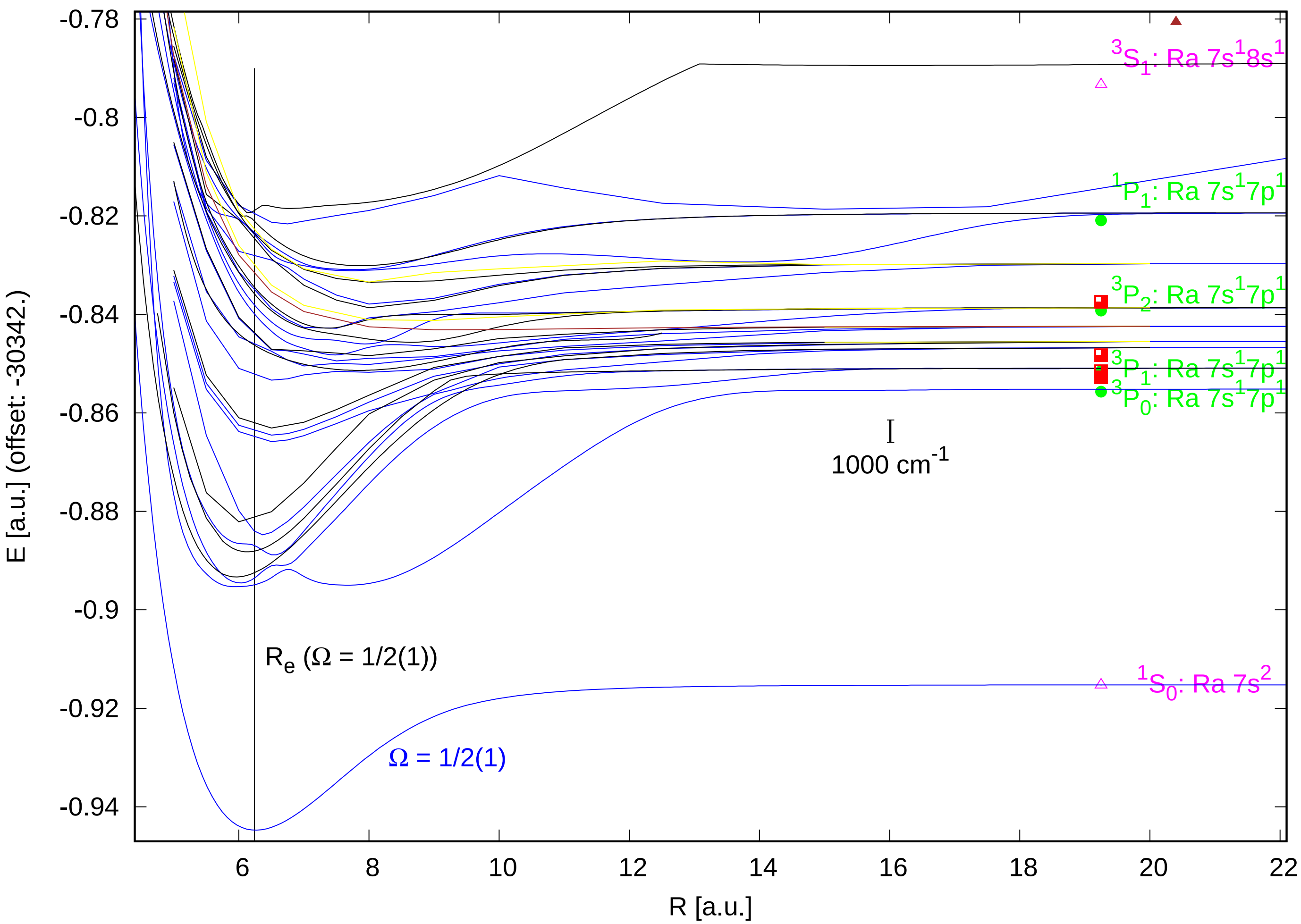


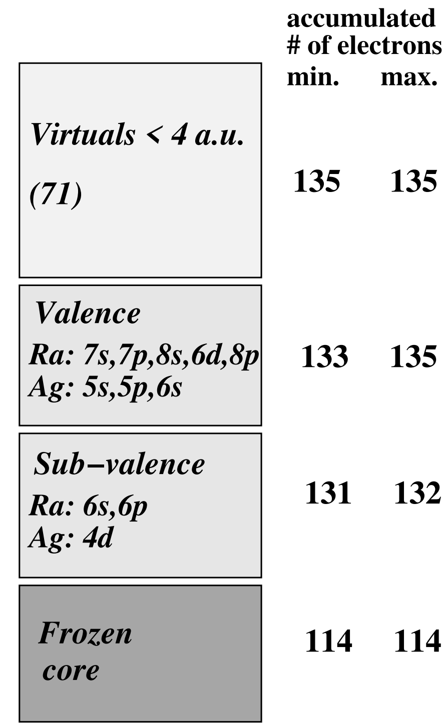
2) Coupled-channel approach

- Does electronic spectrum allow for efficient energy transfer (remove binding energy without heating) ?
- Which states are candidates for photoassociation ?

[26] L. D. Carr, D. DeMille, R. V. Krems, J. Ye, *New J. Phys.* **11** (2009) *055049*

1) RaAg: Complete Spectrum up to $T \approx 5$ eV (TZ basis)





$D_e(1/2(1)) \approx 0.83$ eV

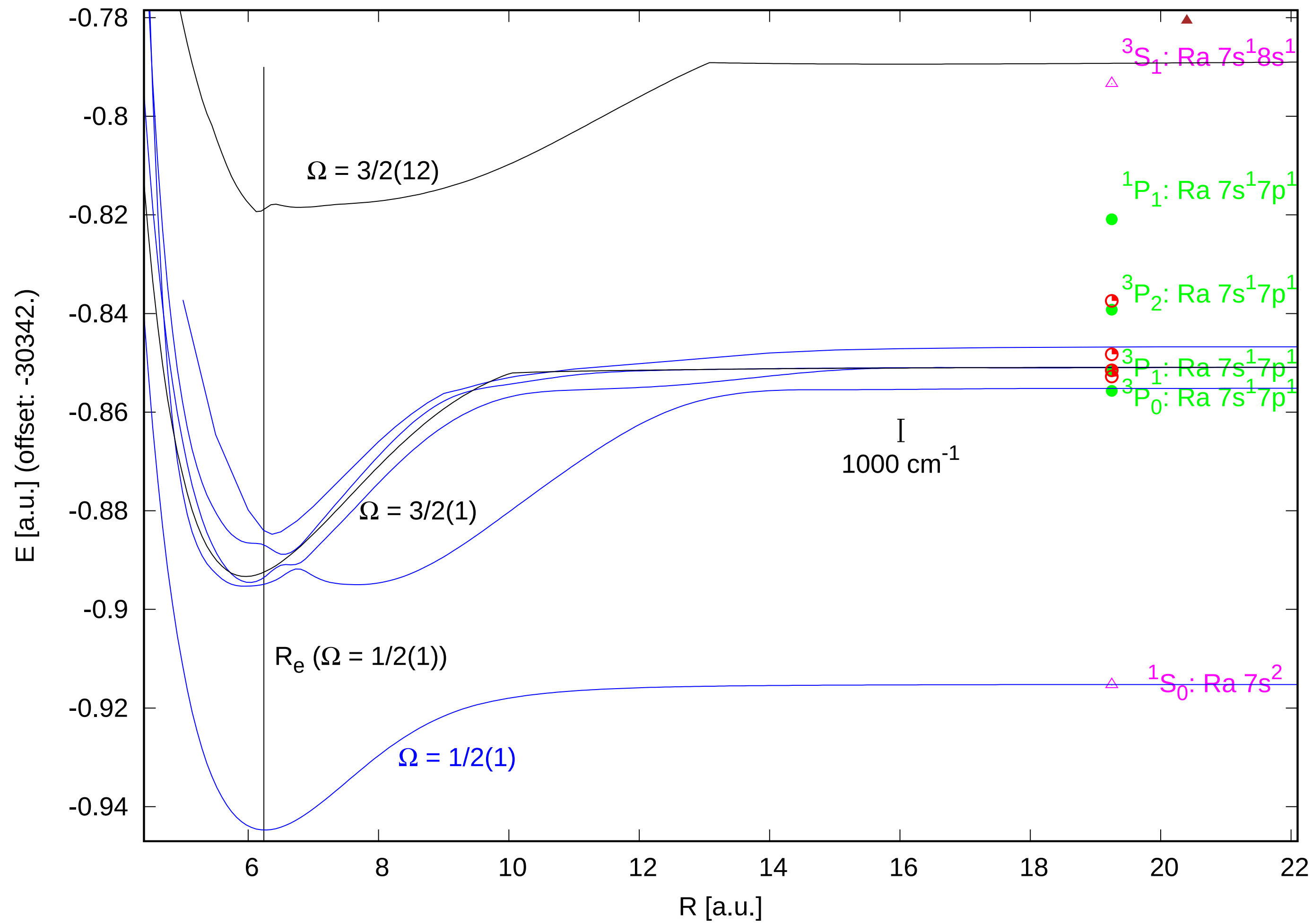
D_e(KRb) ≈ 0.52 eV[26]

D_e(YbRb) ≈ 0.11 eV[27]

[26] S. Kasahara, C. Fujiwara, N. Okada, H. Katô, M. Baba, *J. Chem. Phys.* **111** (1999) *8857*

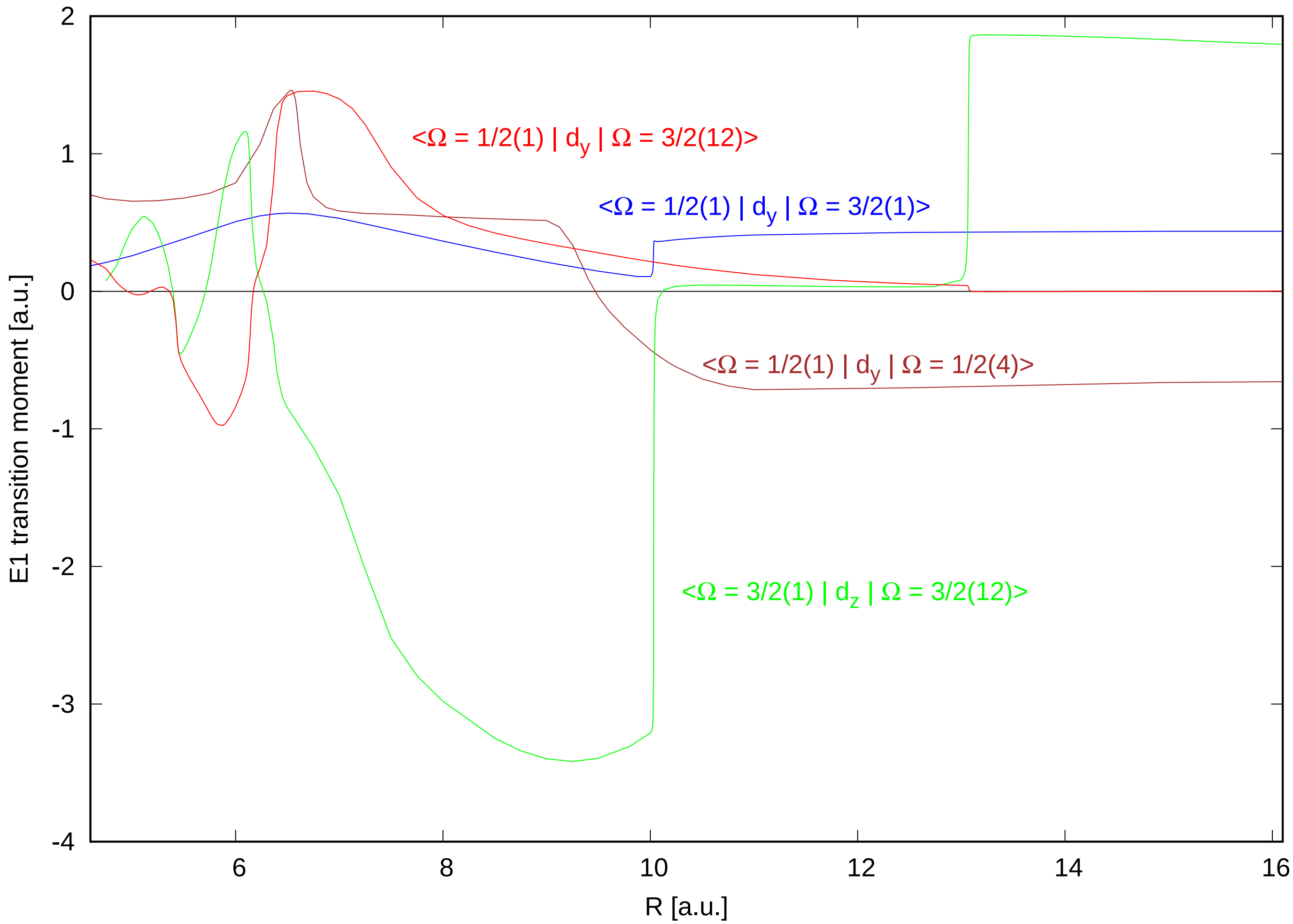
[27] L. K. Sørensen, S. Knecht, T. F., C. M. Marian, *J. Phys. Chem A* **113** (2009) *12607*

1) RaAg: Relevant States $T \approx 5$ eV (TZ basis)



	accumulated # of electrons min.	max.
Virtuals < 4 a.u. *(71)*	**135**	**135**
Valence ***Ra: 7s,7p,8s,6d,8p*** ***Ag: 5s,5p,6s***	**133**	**135**
Sub−valence ***Ra: 6s,6p*** ***Ag: 4d***	**131**	**132**
Frozen core	**114**	**114**

1) RaAg[29]: E1 TDM $d_{XY}(R) = \left\langle \Psi_X \middle| \sum_j q_j \, \hat{\mathbf{r}}_j \middle| \Psi_Y \right\rangle (R)$



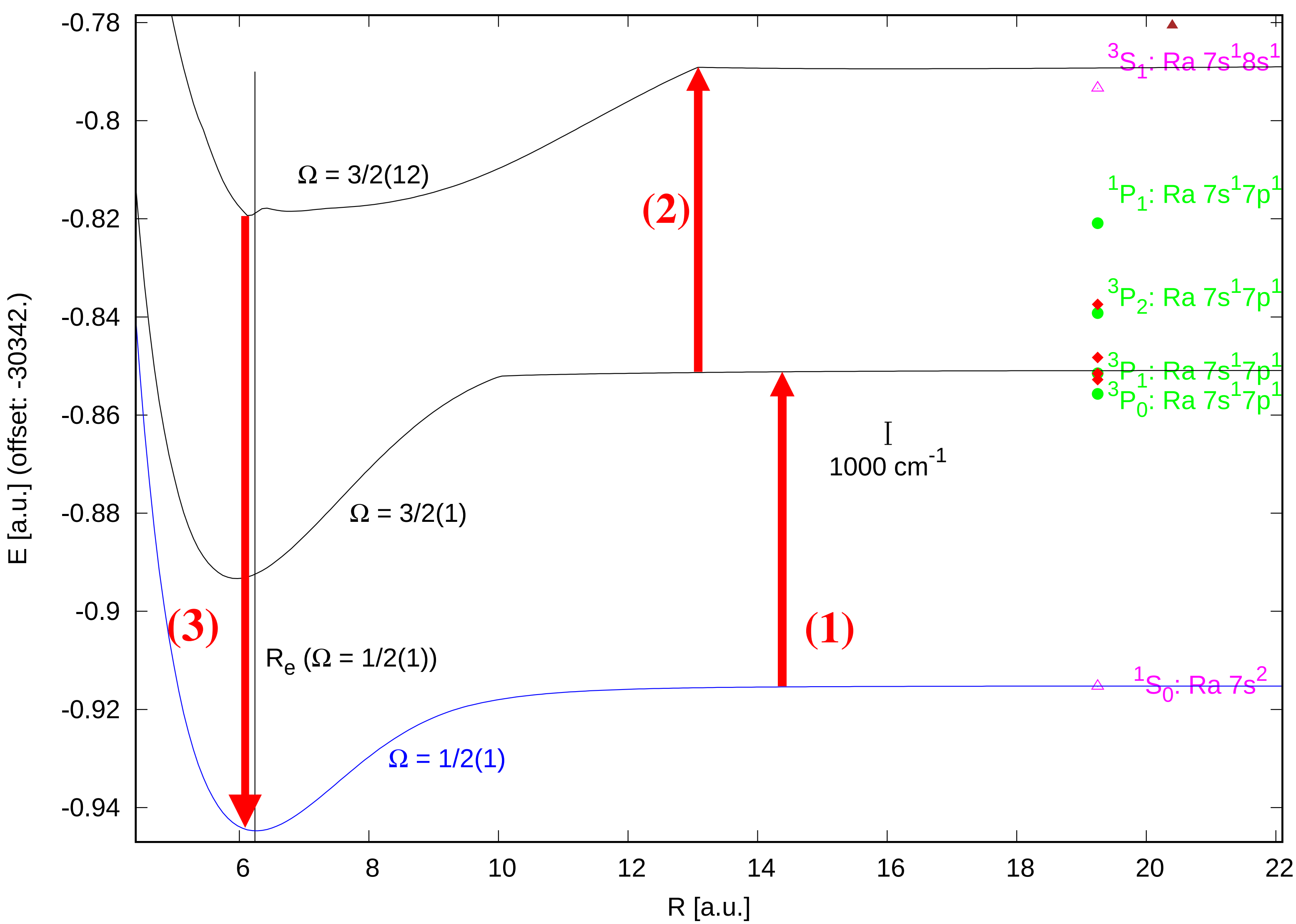
Then: $d_{v,v'} = \int_R \psi_{vX}(R) \, d_{XY}(R) \, \psi_{v'Y}(R) dR$

---

[29] T. Fleig, O. Grasdijk, D. DeMille (2024)

RaAg

1) A Pathway To Assemble RaAg (X) from Trapped Ra-Ag Atom Pairs



Long-Range Theory

Van der Waals interaction potential for two neutral heteronuclear atoms:

$$V(R) = -\frac{C_6}{R^6} - \frac{C_8}{R^8} - \frac{C_{10}}{R^{10}} - \ldots$$

Porsev formalism[30]:

$$C_6(\Omega) = \sum_{j=|J_A-1|}^{J_A+1} \sum_{J=|J_B-1|}^{J_B+1} A_{jJ}(\Omega)\, X_{jJ}$$

with

$$A_{jJ}(\Omega) = \sum_{\mu m M_J} \left\{ (1+\delta_{\mu 0}) \begin{pmatrix} J_A & 1 & j \\ -M_{J_A} & \mu & m_j \end{pmatrix} \begin{pmatrix} J_B & 1 & J \\ -M_{J_B} & -\mu & M_j \end{pmatrix} \right\}^2$$

$$\begin{pmatrix} j_1 & j_2 & j \\ m_{j_1} & m_{j_2} & m_j \end{pmatrix} = \frac{\left\langle j_1 j_2 m_{j_1} m_{j_2} | j_1 j_2 j - m_j \right\rangle}{(-1)^{-j_1+j_2+m_j}\sqrt{2j+1}}$$

$$X_{jJ} = \sum_{\alpha_l, \alpha_k} \frac{\left|\left\langle \alpha_A\, J_A || \hat{T}^{(1)} || \alpha_l\, J_l = j \right\rangle\right|^2 \left|\left\langle \alpha_B\, J_B || \hat{T}^{(1)} || \alpha_k\, J_k = J \right\rangle\right|^2}{E_l - E_A + E_k - E_B}$$

$$\left\langle \alpha\, J || \hat{\boldsymbol{D}} || \alpha'\, J' \right\rangle = \frac{\left|\left|\left\langle \alpha\, J\, M_J | \hat{\boldsymbol{D}} | \alpha'\, J'\, M'_J \right\rangle\right|\right| \sqrt{2J+1}}{\left\langle J'\, 1\, M'_J\, q | J'\, 1\, J\, M_J \right\rangle}$$

[30] S. G. Porsev, M. S. Safronova, A. Derevianko, and C. W. Clark, *Phys. Rev. A* **89** (2014) *022703*

Long-Range Interactions

E1 Transitions in Earth-Alkaline and Alkali Test Systems

$\left|\left\langle X\left(J=\frac{1}{2}\right)||\hat{\boldsymbol{D}}||l\left(J=\frac{1}{2}\right)\right\rangle\right|$ and $\left|\left\langle X\left(J=\frac{1}{2}\right)||\hat{\boldsymbol{D}}||l\left(J=\frac{3}{2}\right)\right\rangle\right|$ in [a.u.]

Li	present			experiment [30]	literature
Excited state	RME	$\Delta\varepsilon$ [cm^{-1}]	f	$\Delta\varepsilon$ [cm^{-1}]	f
$^2P_{1/2}(2p^1)$	3.3197	14909	0.2495	14903.66	
$^2P_{3/2}(2p^1)$	4.6948	14910	0.4991	14904.00	$0.7470\,(^2P)$ [31]
$^2P_{1/2}(3p^1)$	0.1794	30916	0.0015	30925.38	
$^2P_{3/2}(3p^1)$	0.2536	30917	0.0030	30925.38	$0.00482\,(^2P)$ [32]

$\left|\left\langle X(J=0)||\hat{\boldsymbol{D}}||l(J=1)\right\rangle\right|$ [a.u.]

Be	present			experiment [30]	literature
excited state	RME	$\Delta\varepsilon$ [cm^{-1}]	f	$\Delta\varepsilon$ [cm^{-1}]	f
$^3P_1(2s^12p^1)$	0.0002	21977	0.0000	21978.93	
$^1P_1(2s^12p^1)$	3.2615	42585	1.3760	42565.35	1.374 [33]
$^1P_1(2s^13p^1)$	0.2111	60347	0.0082	60187.34	0.0086 [33]

[30] A. Kramida, Yu. Ralchenko, J. Reader, and and NIST ASD Team, *NIST Atomic Spectra Database* (2019)

[31] Z.-C. Yan, M. Tambasco, and G. W. F. Drake, *Phys. Rev. A* **57** (1998) *1652*

[32] L. Qu, Z. Wang, and B. Li, *Eur. Phys. J. D* **5** (1999) *173*

[33] S. Nasiri, L. Adamowicz, and S. Bubin, *J. Phys. Chem. Ref. Data* **50** (2021) *043107*

Long-Range Interactions

Earth-Alkali Atoms

$$\left|\left\langle X(J=0)||\hat{\boldsymbol{D}}||l(J=1)\right\rangle\right| \text{ [a.u.]}$$

Ca		present			experiment		
state	CI model	RME	$\Delta\varepsilon$ [cm^{-1}]	f	RME	$\Delta\varepsilon$ [cm^{-1}] [34]	f
$^1P_1(4p^1)$(3)	SDTQ_SD	4.98	25200	1.90	4.912^*	23652.304	$1.7332(7)$[35]
$^1P_1(5p^1)$(10)	SDTQ_SD	0.23	43000	0.01		36731.615	
$^1P_1(6p^1)$(17)	SDTQ_SD	0.93	52400	0.14		41679.008	

Dispersion coefficients for RaAg valence-isoelectronic systems:

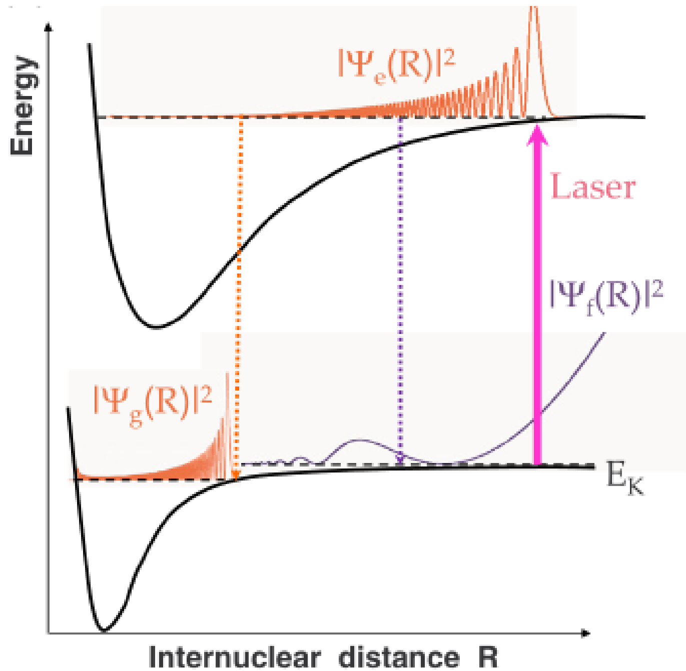
	C_6 [a.u.]	
System	present	literature
BeLi $X^2\Sigma_{1/2}$	464	478 a[35]
CaLi $X^2\Sigma_{1/2}$	1581 1644^*	1689
CaCa $X(\Omega=0)$	2030^*	$2080(7)$ b[35]

[34] A. Kramida, Yu. Ralchenko, J. Reader, and and NIST ASD Team, *NIST Atomic Spectra Database* (2021)

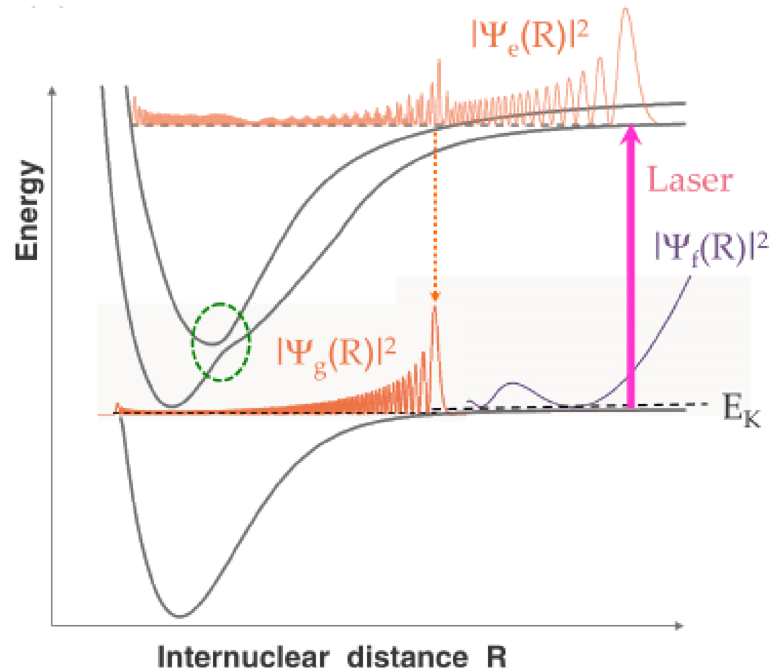
[35] (a) J. Jiang, Y. Cheng, and J. Mitroy, *J. Phys. B: At. Mol. Opt. Phys.* **46** (2013) *134305*
(b) O. Allard, C. Samuelis, A. Pashov, H. Knöckel, and E. Tiemann, *Eur. Phys. J. D* **26** (2003) *155*

“Building” RaAg in a DLT EDM Experiment

- Photoassociating ultracold atoms into ultracold molecules[37]



1) Direct approach



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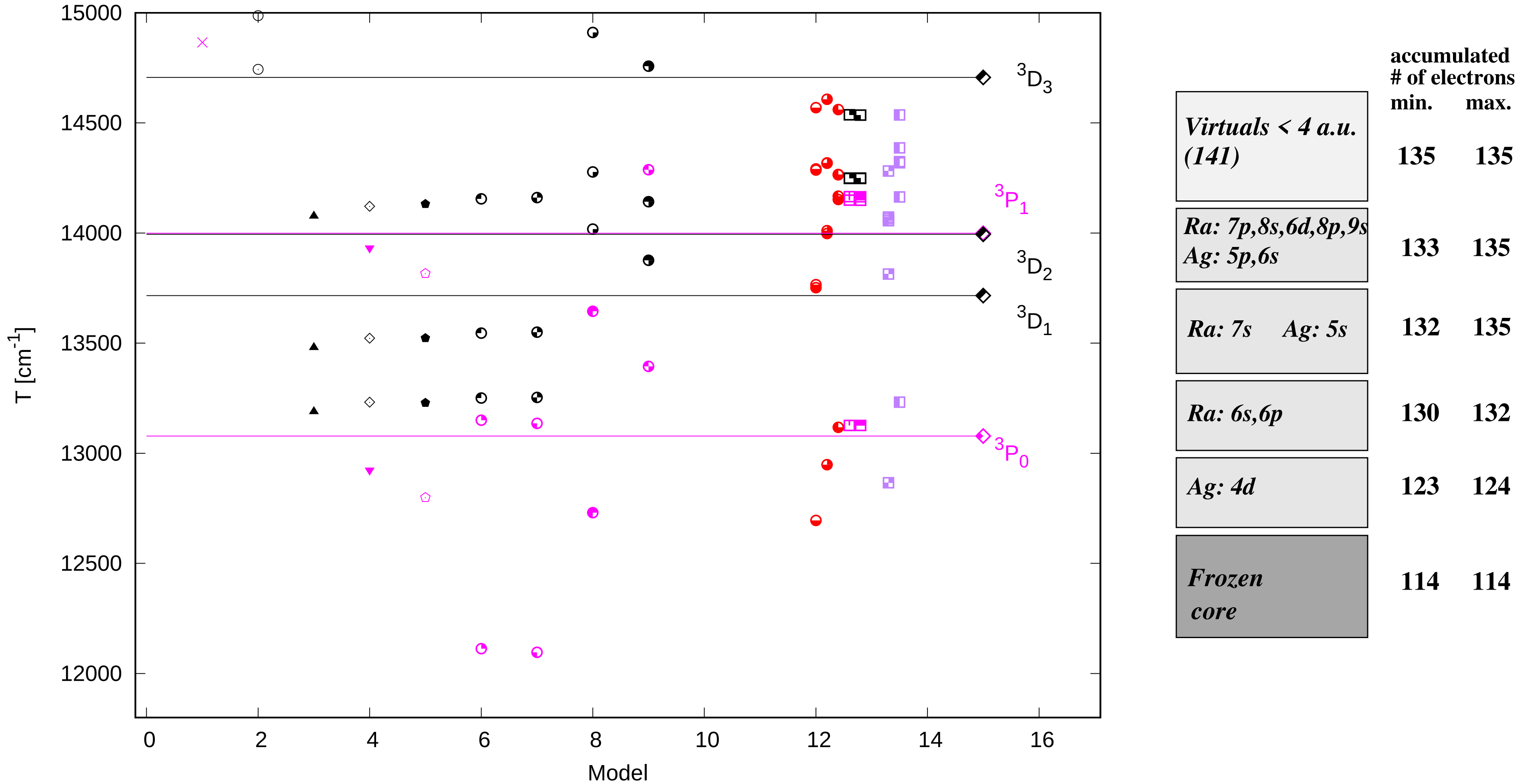
- Which states are candidates for photoassociation ?

[37] L. D. Carr, D. DeMille, R. V. Krems, J. Ye, *New J. Phys.* **11** (2009) *055049*

2) RaAg: Limited Spectrum up to $T \approx 3$ eV (QZ basis)



4: SD8_8s6d9s7d7p8pSDT_SD10

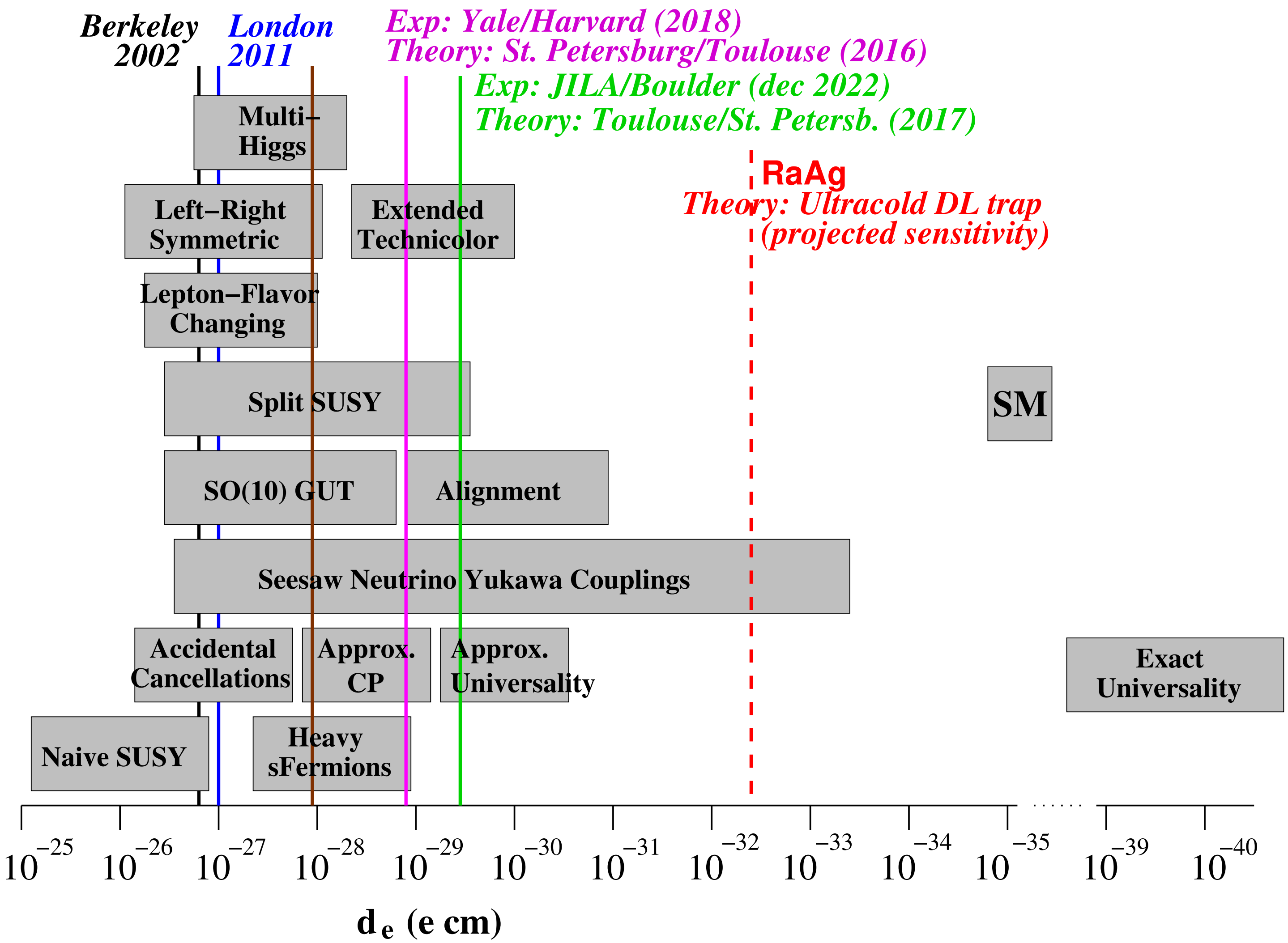
9: SD8_8s6d9s7d7p8pSDTQ_SD10

12: RaAg (SDT)

13: RaAg (SDTQ)

eEDM Constraint on Beyond-Standard-Model Theories

Single-source interpretation (21??)



EDM Science

- Electron EDM interactions (HfF^+, ThO, Hg, Tl, TaO^+, RaAg et al.)

  T. F., D. DeMille, *New J. Phys.* **23** (2021) *113039*

  T. F., L. V. Skripnikov, *Symmetry* **12** (2020) *498*

  T. F., M. Jung, *J High Energy Phys. (JHEP)* **07** (2018) *012*

  T. F., *Phys. Rev. A* **96** (2017) *040502(R)*

  T. F., *Phys. Rev. A* **95** (2017) *022504*

  M. Denis, T. F., *J. Chem. Phys.* **145** (2016) *214307*

- Nuclear Schiff-moment interactions (Xe, Hg, TlF, FrAg et al.)

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  M. Hubert, T. F., *Phys. Rev. A* **106** (2022) *022817*

- Weak neutral current interactions (Xe, Hg, Ra, TlF)

  T. F., *Phys. Rev. A* **109** (2024) *022807*

  T. F., *Phys. Rev. A* **99** (2019) *012515*

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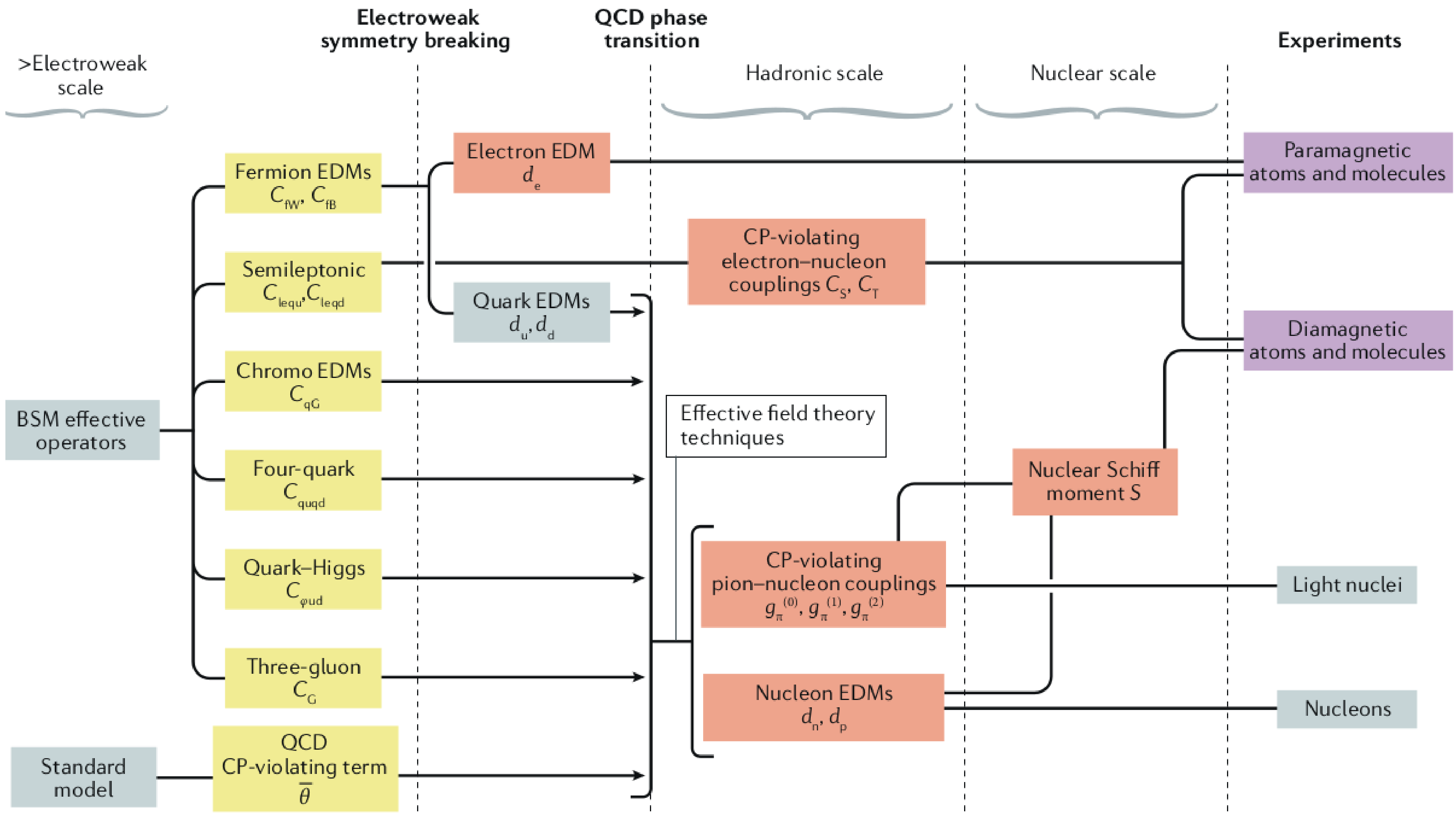
  T. F., M. K. Nayak, M. G. Kozlov, *Phys. Rev. A* **93** (2016) *012505*

EDMs and their possible sources: An overview



W. Cairncross, J. Ye, *Nat. Rev. Phys.* **1** (2019) *510*

Tensor-Pseudotensor $\mathcal{P},\mathcal{T}$-odd Nucleon-Electron Interaction

Effective Hamiltonian for a single electron[37] for Ne neutral weak current

$$\hat{H}^{\text{eff}}_{\text{T-PT-ne}} = \frac{\imath G_F}{\sqrt{2}} \sum_N C_T^N \, \rho_N(\mathbf{r}) \gamma^0 \sigma_{N\,\mu\nu} \gamma^5 \sigma^{\mu\nu}$$

Using the identity

$$\sigma_{N\,\mu\nu} \gamma^5 \sigma^{\mu\nu} = 2\gamma_N^0 \boldsymbol{\gamma}_N \cdot \boldsymbol{\Sigma} + 2\gamma^0 \boldsymbol{\Sigma}_N \cdot \boldsymbol{\gamma}$$

and $\langle\psi|\boldsymbol{\Sigma}|\psi\rangle = 0$ for closed-shell systems:

$$\hat{H}^{\text{eff}}_{\text{T-PT-ne}} = \frac{\imath G_F}{\sqrt{2}} \sum_N 2C_N^T \, \boldsymbol{\Sigma}_N \cdot \boldsymbol{\gamma} \rho_N(\mathbf{r})$$

For nuclear state $|I, M_I = I\rangle$ isotope-specific many-electron Hamiltonian[38]:

$$\hat{H}^{\text{eff}}_{\text{T-PT-ne}} = \imath\sqrt{2} G_F \, C_T^A \, \langle\Sigma\rangle_A \sum_{j=1}^{n} (\gamma_3)_j \, \rho(\mathbf{r}_j)$$

[37] K. Yanase, N. Yoshinaga, K. Higashiyama, N. Yamanaka *Phys. Rev. D* **99** (2019) *075021*
[38] T. F., M. Jung *Phys. Rev. A* **103** (2021) *012807*

Molecular T-PT-ne Interaction Constant[40]

Energy shift of state E in a molecule:

$$\Delta\varepsilon_E = \left\langle \psi_E^{(0)} \middle| \hat{H}^{\text{eff}}_{\text{T-PT-ne}} \middle| \psi_E^{(0)} \right\rangle = W_T \, C_T^A$$

It then follows that

$$W_T(X) = \sqrt{2} G_F \langle \Sigma \rangle_A \left\langle \psi_E^{(0)} \middle| \imath \sum_{j=1}^{n} (\gamma_3)_j \, \rho_X(\mathbf{r}_j) \middle| \psi_E^{(0)} \right\rangle$$

In atoms

$$d_a = C_T^A \, \alpha_{C_T}$$

where

$$\alpha_{C_T} := \frac{\left\langle \hat{H}^{\text{eff}}_{\text{T-PT-ne}} \right\rangle_{\psi^{(0)}(E_{\text{ext}})}}{E_{\text{ext}}}$$

[40] T. F., *Phys. Rev. A* **109** (2024) *022807*

W_T(Tl) in TlF($^1\Sigma_0$) from Hartree-Fock theory[41]

model	W_T(Tl) [kHz $\langle\Sigma\rangle_A$]	total energy
Hartree-Fock[41]	-1.34	
Hartree-Fock[42]	-0.851	
Dirac-Coulomb HF[43]	-4.641	-20374.4108
cGHF-ZORA-wr[44]	-4.690	
DZ/DCHF	-4.601	-20374.41122770
TZ/DCHF	-4.673	-20374.46576781
QZ/DCHF	-4.684	-20374.47704191
QZ+dens+sp/DCHF	-4.684	-20374.47660904

[41] T. F., *Phys. Rev. A* **109** (2024) *022807*

[41] Converted EDM in terms of C_T from E. A. Hinds, C. E. Loving, and P. G. H. Sandars, *Phys. Lett. B* **62** (1976) *97* using the external electric field and interaction constant reported *ibidem*

[42] Value reported in D. Cho, K. Sangster, and E. A. Hinds, *Phys. Rev. A* **44** (1991) *2783* which is the corrected result from Ref. P. V. Coveney and P. G. H. Sandars, *J. Phys. B: At. Mol. Opt. Phys.* **16** (1983) *3727*

[43] H. M. Quiney, J. K. Lærdahl, T. Saue, and K. Fægri Jr., *Phys. Rev. A* **57** (1998) *920*

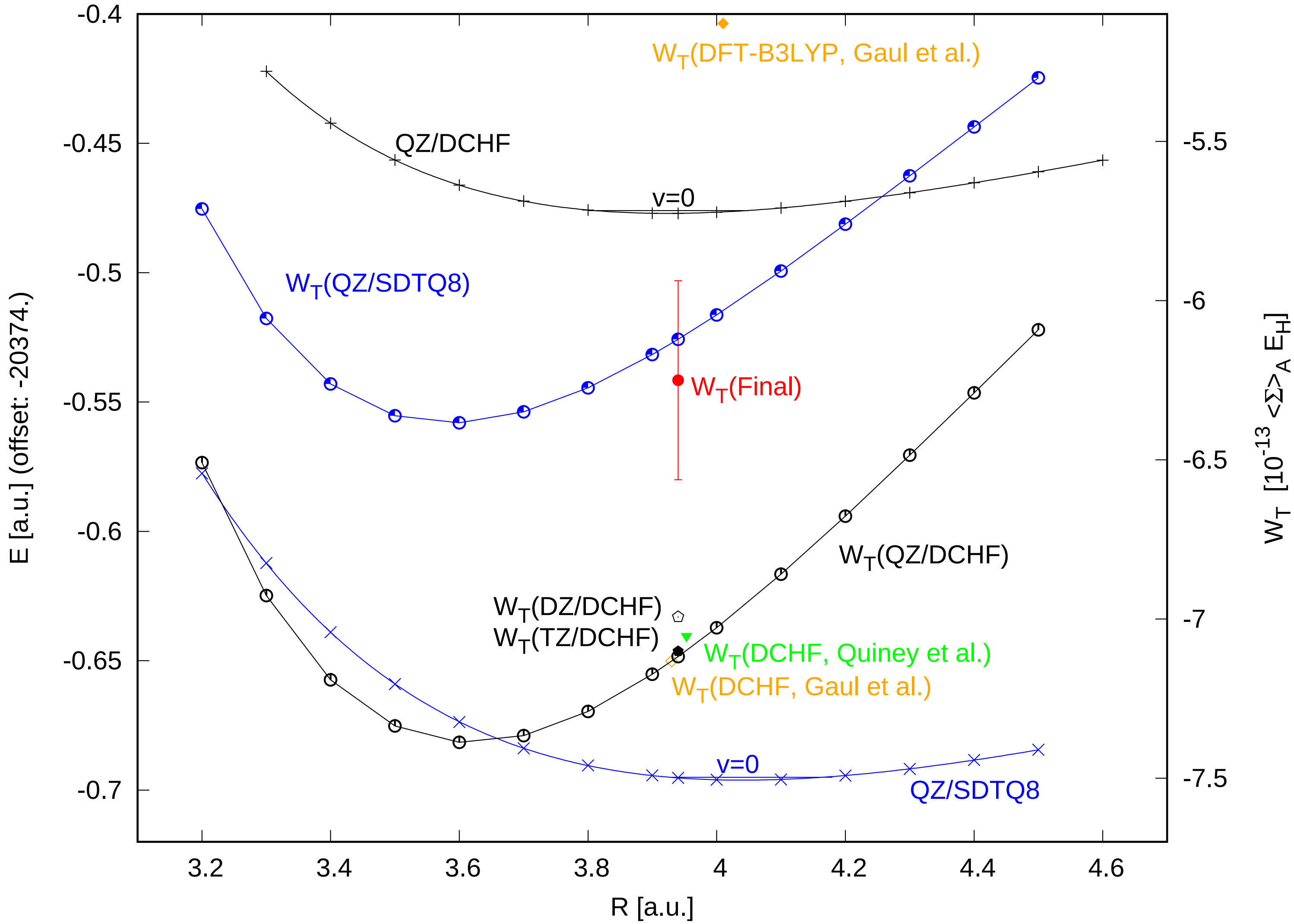
[44] Value from Ref. K. Gaul, R. Berger, *J. Chem. Phys.* **152** (2020) 044101 with adapted sign, $R = 3.93$ a.u.

W_T(Tl) in TlF($^1\Sigma_0$) from GAS-CI[46]

model	W_T(Tl) $[10^{-13}\langle\Sigma\rangle_A$ *a.u.*$]$	$[\text{kHz}\ \langle\Sigma\rangle_A]$	total energy
QZ/DCHF	-7.12	-4.68	-20374.47704191
QZ/SD4_6.5au	-6.40	-4.21	-20374.53183162
QZ/SDTQ4_6.5au	-6.33	-4.17	-20374.53321310
QZ/S4_SDTQ8_6.5au	-6.08	-4.00	-20374.61641168
QZ/S4_SDTQQ8_6.5au	-6.07	-4.00	-20374.61647310
QZ/SD8_6.5au	-6.47	-4.26	-20374.67877868
QZ/SDTQ8_6.5au	-6.12	-4.03	-20374.69523546
QZ/SD18_6.5au	-6.58	-4.33	-20374.98617776
QZ/SD20_6.5au	-6.61	-4.35	-20375.05000858
QZ/SD28_6.5au	-6.49	-4.27	-20375.17819744
QZ/SD28_18au	-6.59	-4.33	-20375.37322100
QZ/SD36_18au	-6.59	-4.34	-20375.38490094
Final	**-6.25**	$\mathbf{-4.11}$	

[46] T. F., *Phys. Rev. A* **109** (2024) *022807*

W_T(Tl) in TlF($^1\Sigma_0$) from Correlated Theory[47]



[47] T. F., *Phys. Rev. A* **109** (2024) *022807*

Schiff-Moment Interactions and Method Development

Mickaël Hubert, Lecturer

Schiff-moment interaction implementation

Basis sets

Aurélien Marc, PhD Student

Schiff-moment interaction calculations in molecules

DCG-GASCI

Thanks for your attention !